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Disclaimer

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Abstract

The CAMBRIC nuclear test was conducted beneath Frenchman Flat at the Nevada Test Site on May 14, 1965. The nuclear device was emplaced in heterogeneous alluvium, approximately 70 m beneath the ambient water table, which is itself 220 m beneath the ground surface. Approximately 10 years later, groundwater adjacent to the test was pumped steadily for 16 years to elicit information on the migration of residual radionuclide migration through the saturated zone. The pumping well effluent – containing mostly soluble radionuclides such as tritium, ^{14}C, ^{36}Cl, ^{85}Kr, ^{129}I, and ^{106}Ru – was monitored, discharged to an unlined ditch, and allowed to flow towards Frenchman Lake over one kilometer away. Discharged water and radionuclides infiltrated into the ground and created an unexpected second experiment in which the migration of the effluent through the unsaturated zone back to the water table could be studied. In this paper, the pumping and effluent data are being utilized in conjunction with a series of geologic data, new radionuclide measurements, isotopic age-dating estimates, and vadose zone flow and transport models to better understand the movement of radionuclides between the ditch and the water table. Measurements of radionuclide concentrations in water samples produced from a water table monitoring well 100 m away from the ditch indicate rising levels of tritium since 1993. The detection of tritium in the monitoring well occurs approximately 16 years after its initial discharge into the ditch. Modeling and tritium age dating have suggested 3 to 5 years of this 16-year transit time occurred solely in the vadose zone. They also suggest considerable recirculation of the pumping well discharge back into the original pumping well. Notably, there have been no observations of ^{14}C or ^{85}Kr at the water table, suggesting their preferential retention or volatilization during transit to the water table. Overall, the long term nature of the experiment, the variety of chemical measurements and isotopic interpretations, and their incorporation into a unified modeling analysis, have contributed to a unique perspective for interpreting radionuclide migration in a deep unsaturated system.

1. Introduction

Contaminant migration in deep unsaturated environments is of considerable concern across much of the US Department of Energy complex, and the scientific and technical challenges for addressing many of these problems are far from being solved (USDOE, 2001). Vadose zone contamination at the Hanford Reservation in Washington is often cited as a critical example (e.g., USDOE, 1999). The Nevada Test Site and environs offer additional examples (Figure 1), most notably at the proposed Yucca Mountain nuclear waste repository, adjacent to NTS, as well as on the NTS proper, where approximately 2/3 of the 908 underground nuclear tests were conducted above the water table (USDOE, 1977). Many components of the residual radioactive inventory associated with these tests (e.g., Smith et al., 2003) can behave as mobile tracers indicative of water, gas phase, and particulate transport in the subsurface. Most of these tracers continue to be useful to this day, long after the testing program has ceased. This paper deals with one such test and summarizes recent work that takes advantage of what has now become a 40-year-long experiment involving infiltration and contaminant migration in a deep vadose zone system.

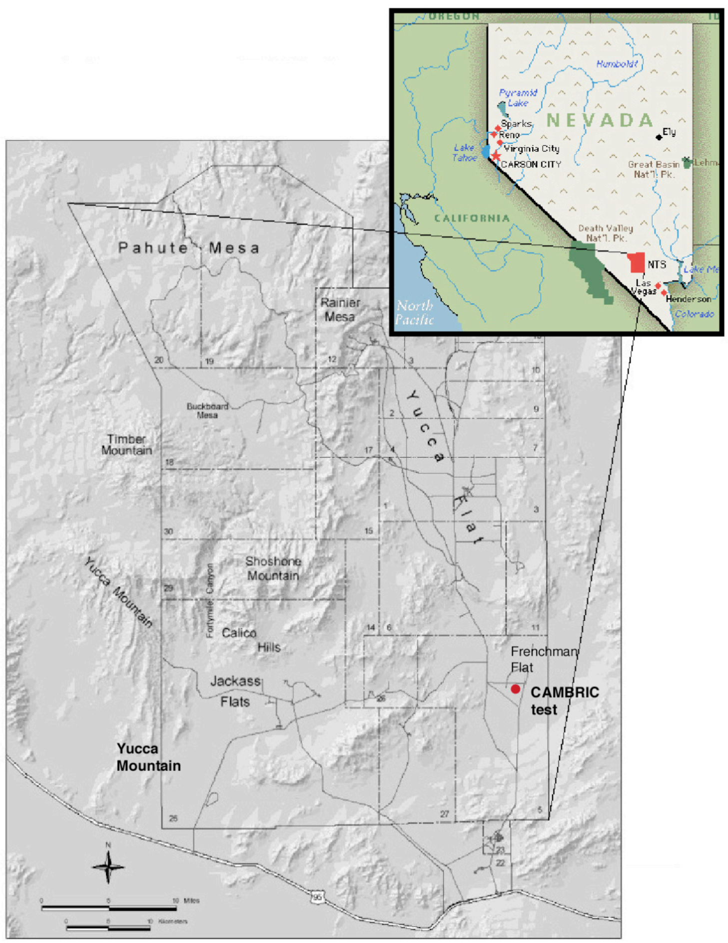

Figure 1: Map of the Nevada Test Site and its location within Nevada. The CAMBRIC test was conducted in Area 5 in Frenchman Flat, in the southeast portion of the test site. Yucca Mountain is in the southwest portion of the test site.

2. The CAMBRIC Test and Related Experiments

The CAMBRIC nuclear test was conducted beneath Frenchman Flat at the Nevada Test Site (NTS) on May 14, 1965. Frenchman Flat is located in Area 5 near the southeast corner of the NTS, as shown in Figure 1. The test device was positioned in heterogeneous alluvium, 294 m beneath the ground surface, and approximately 74 m beneath the ambient water table. The announced energy-equivalent yield of the test was 0.75 kilotons. The explosion created a detonation cavity approximately 22 m in diameter that was subsequently filled in by collapsed alluvium and saturated with groundwater (Hoffman et al., 1977; Bryant, 1992). The collapsed "chimney" did not extend as far as the ground surface as there was no observable crater.

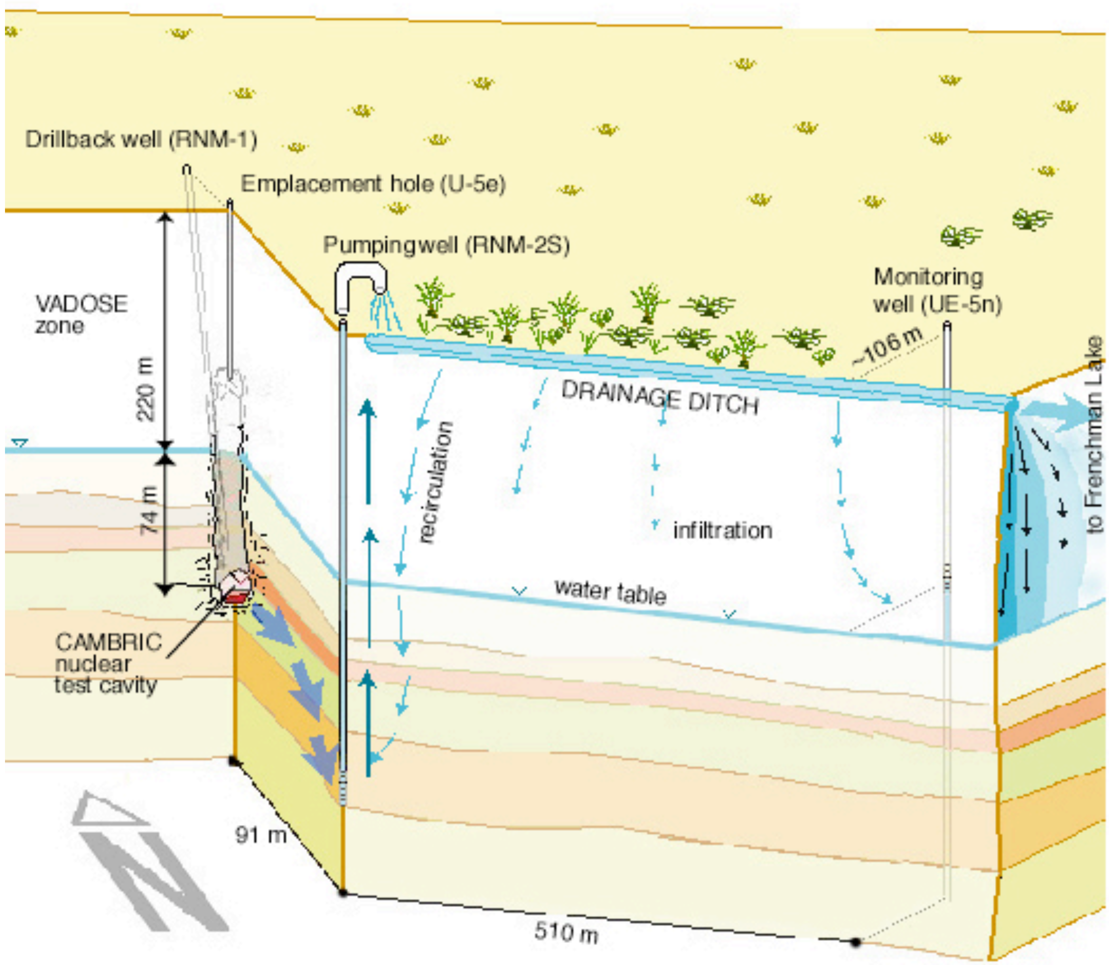

Figure 2: Schematic of the CAMBRIC pumping experiment in Frenchman Flat at the Nevada Test Site, showing the test emplacement hole (U-5e), cavity and collapsed chimney, pumping well RNM-2S, drainage ditch, and monitoring well UE-5n.

2.1 The Groundwater Pumping Experiment.

The CAMBRIC test is relatively unique at the NTS in that it has been extensively characterized and monitored for post-test environmental impacts. Beginning in October 1975, approximately 10 years after the test, groundwater adjacent to the test cavity was pumped steadily for 16 years, with a few short interruptions, in order to elicit information on test-related radionuclide migration in the saturated zone (Hoffman, et al, 1977; Bryant, 1992). Figure 2 illustrates the configuration of the original CAMBRIC device emplacement hole (U-5e), the diagnostic, post-test drillback well (RNM-1), the experimental pumping well (RNM-2S) and a nearby monitoring well (UE-5n) in the saturated zone. As shown in this schematic, the pumping well (RNM-2S) was located approximately 91 m south of the emplacement hole (U-5e), and was screened over an approximately 25 m interval between depths of 318 and 343 m, slightly below the test cavity.

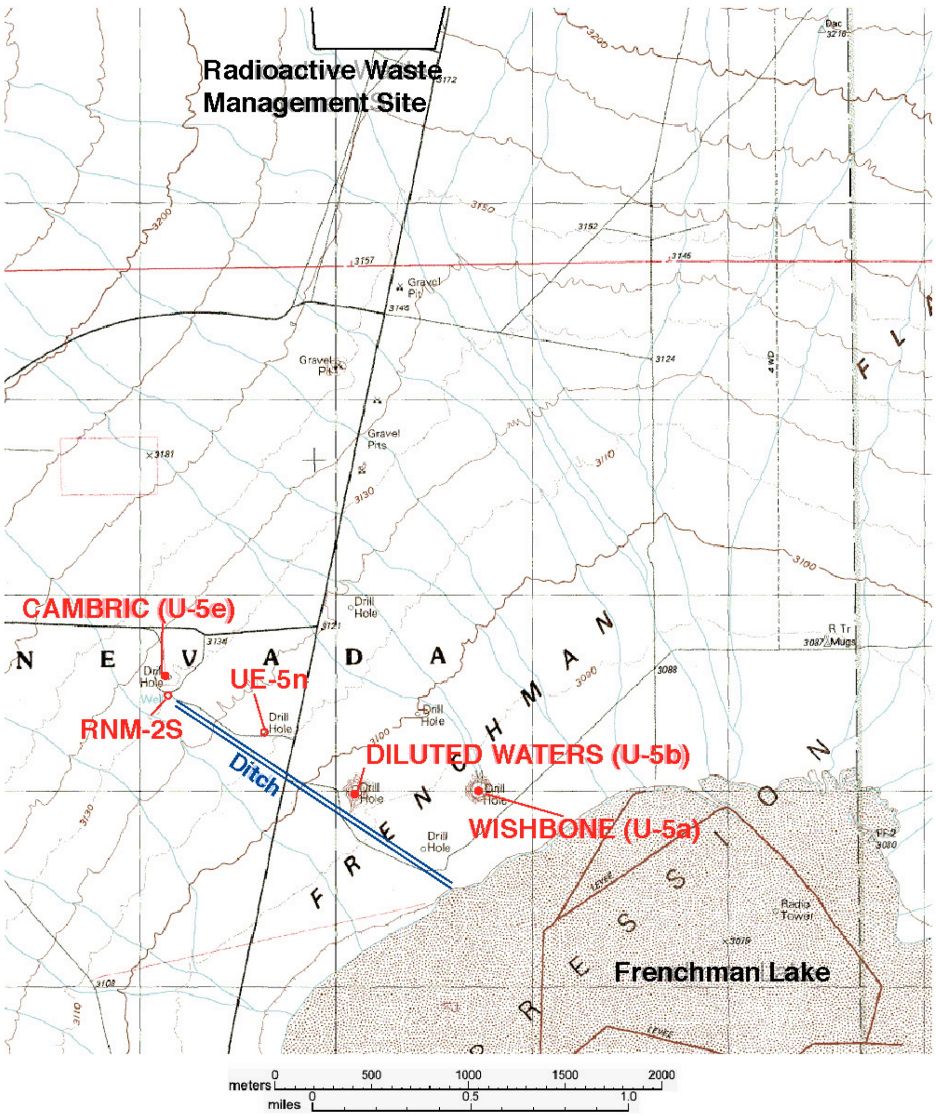

Figure 3: Map of the Frenchman Flat area at the Nevada Test Site showing the CAMBRIC test location and wells RNM-2S and UE-5n associated with the pumping experiment. The effluent ditch and nearby emplacement locations of the WISHBONE and DILUTED WATERS tests (and associated surface craters) are also shown. The Radioactive Waste Management Site is at the top.

The pumping well effluent was regularly monitored for its radionuclide content. The effluent was discharged to an unlined ditch and allowed to flow towards the Frenchman Lake playa, approximately 1.6 km to the southeast. Figure 3 shows a map of the region surrounding the experiment, including the effluent ditch and the emplacement locations of two other underground nuclear tests (DILUTED WATERS, U-5b, and WISHBONE, U-5a). Over the 16 years of the experiment, extensive growth of saltcedar and cattails, both invasive nonnative shrubs (Brock, 1994), developed along the ditch.

Radionuclides regularly observed in the pumping well effluent included tritium (^{3}H or T, occurring in molecular water as HTO), ^{36}Cl, ^{85}Kr, and ^{129}I (Bryant, 1992). Sporadic observations of ^{106}Ru and ^{99}Tc were also made. Figure 4 shows the tritium activity and ^{36}Cl concentration measured in the effluent between the start of pumping in October 1975 until its cessation in the fall of 1991 (all data are decay corrected to May 14, 1965). The recovery curves appear to reflect relatively "complete" breakthrough profiles for these radionuclides, which are generally quite mobile in groundwater. Similar behavior was observed for ^{85}Kr and ^{129}I (not shown; see Bryant, 1992). Relatively "immobile" species such as ^{90}Sr, ^{137}Cs, and $^{238,239}Pu$ were looked for, but never detected (Bryant, 1992). Detection of other radionuclides was not formally pursued during the pumping test, although test-related ^{14}C was later identified in the pumping well in 2000 as part of the current work, indicating its apparent co-migration with other mobile radionuclides pumped and discharged into the ditch.

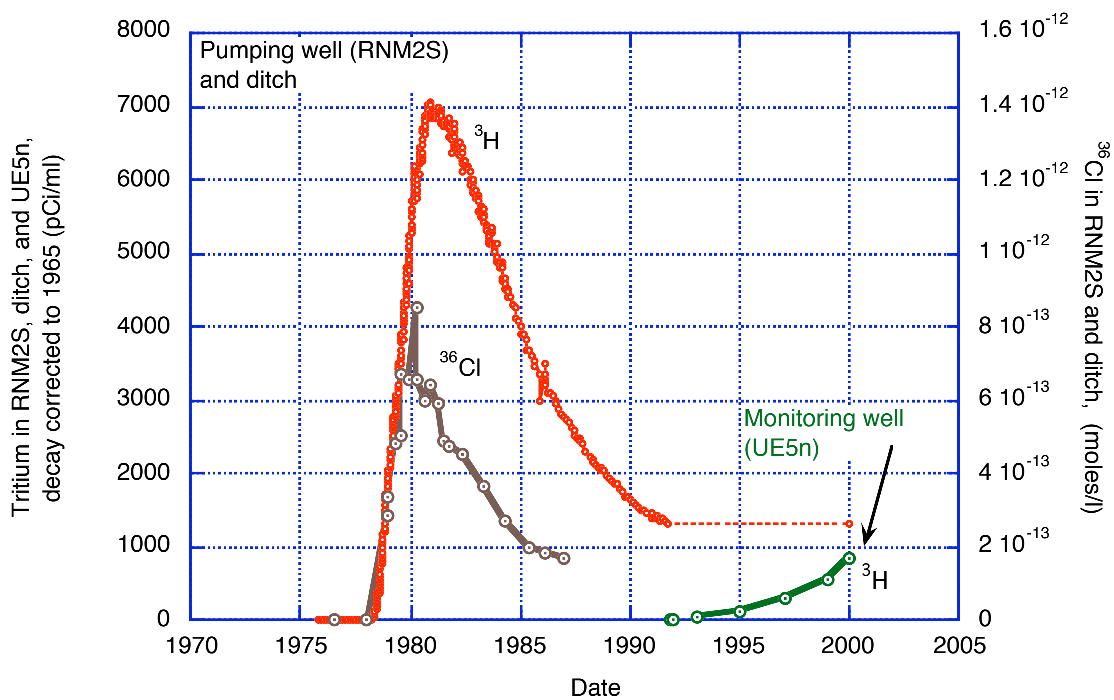

Figure 4: Tritium (^{3}H) activity and ^{36}Cl concentrations observed in the pumping well (RNM-2S) between the initiation (1975) and cessation of pumping (1991), and in an isolated measurement made in 2000. Rising tritium activity in monitoring well UE-5n, beginning between 1991 and 1994, is also shown. All data are decay corrected to their values on May 14, 1965.

As shown in Figure 4, the first arrivals of tritium and ^{36}Cl were not observed until more than two years (900 days) of pumping, and this occurred after the initial pumping rate (300 gpm, or 1635 m^3/d) was doubled (to 600 gpm, or 3270 m^3/d) after 700 days. The first arrival times for ^{85}Kr and ^{129}I (not shown) were generally similar. The peak tritium concentration was observed in 1981. The peak concentration of ^{36}Cl occurred somewhat earlier, while peak concentrations of ^{85}Kr and ^{129}I (not shown) were substantially delayed. Measured concentrations of ^{106}Ru and ^{99}Tc were too infrequent to allow similar comparisons. In general, differences in the shapes of these profiles can been attributed to detonation effects that affect the initial distribution of residual radioactivity (Bryant, 1992; Guell and Hunt, 2003; Tompson et al., 2002), as well as slight differences in the overall mobility of the radionuclides in groundwater. Extensive analyses and studies of the radionuclide migration between the CAMBRIC cavity and the pumping well were conducted both during and after this experiment (e.g., Hoffman, et al., 1977; Burbey and Wheatcraft, 1986; Ogard et al., 1988; Bryant, 1992; Tompson et al., 1999a; and Guell and Hunt 2003).

2.2 The Serendipitous Infiltration Experiment.

It is notable that there was, initially, little interest in the fate of radionuclides once they were discharged into the ditch. A common perception at the time was that infiltration of contaminated ditch effluent into the 220 meters of unsaturated alluvium would not constitute a significant threat with regard to recontamination of groundwater. This concern was challenged in the mid 1980s and early 1990s with a series of soil moisture and water quality measurements in which the infiltration of water and migration of tritium and ^{36}Cl were measured horizontally to about 7 m and vertically to at least 30 m beneath and away from the ditch (Buddemeier et al., 1991, Ross and Wheatcraft, 1994).

In the 1980s, a series of flume-based measurements of the ditch flow rate between the pumping well and Frenchman Lake indicated that a loss of approximately 175 gpm (or 954 m^3/d, roughly one third of the upstream flow rate) occurred over an approximately 1000-m distance (Bryant, 1992). This loss amounts to approximately 0.01 kg-water/s per meter of ditch length (yet was estimated by Ross and Wheatcraft, 1994, to be closer to 0.02 kg-water/s per meter of ditch length). Small diurnal fluctuations in these flow rates have also been observed and attributed to daily transpiration behavior in the saltcedar and cattails. Based upon these observations, it had been suggested that infiltrated water (and tritium) could reach the water table in approximately 9 years (Bryant, 1992).

Between 1991 and 1993, regular tests of groundwater in monitoring well UE–5n, located 500 m away from the pumping well and 106 m perpendicular to the ditch (Figures 2 and 3), began to show rising levels of tritium at the water table, as shown in Figure 4, apparently confirming that infiltrated radionuclides have reached groundwater after transiting the roughly 220 m of unsaturated alluvium (Davisson, et al., 1994). This suggested a 13- to 15-year transit time for tritium to initially move vertically from the ditch to the water table and then horizontally to the monitoring well.

These measurements were initially made on bailed samples taken from the 3-m screened interval of the well, situated just below the water table at a depth of 220 m, and have continued regularly using pumped samples through 2002 (Rose et al., 2002).

3. Additional Analyses of the Infiltration Experiment

The original CAMBRIC pumping experiment, the associated ditch infiltration observations, and the ongoing monitoring of water quality in well UE-5n have created a second, and rather unique, 40-year long experiment involving coupled groundwater-vadose zone flow and transport processes. Aspects of this experiment continue to evolve today, as the shallow soil profile continues to dry out, as vegetation patterns (or other biogeochemical indicators) adjacent to the ditch respond to drier conditions, and as distributions of infiltrated moisture and radionuclides migrate through the deeper portions of the vadose zone and groundwater system.

Below, the pumping and effluent data will be used in conjunction with a series of geologic data, new radionuclide measurements, isotopic age-dating estimates, and vadose zone flow and transport models to better understand the movement of radionuclides between the ditch and the water table. This will offers a basis to more directly quantify these processes, reassess the meaning and importance of the original pumping experiment data, and develop new ideas to study additional aspects of the experiment.

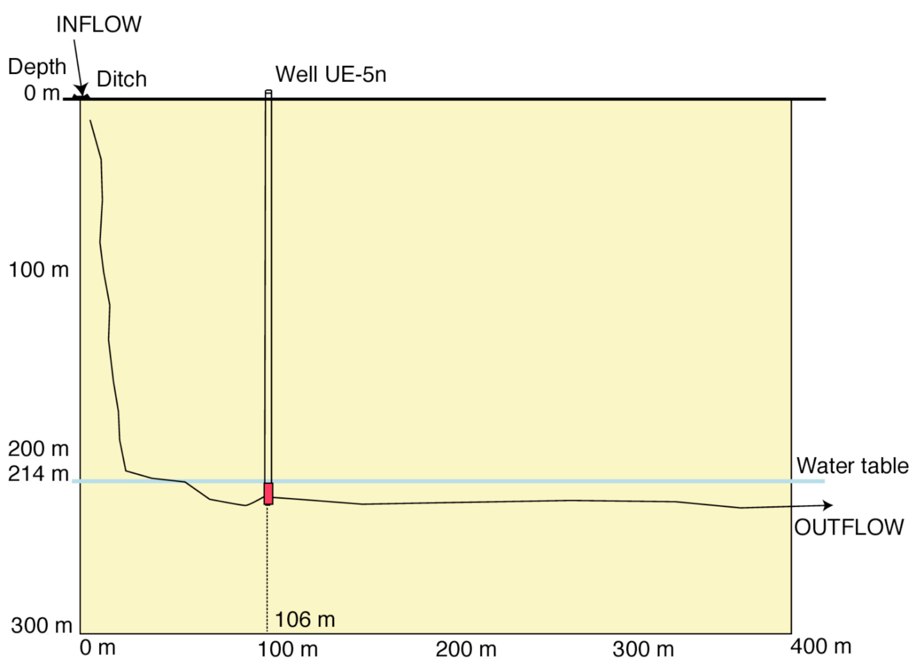

Figure 5: Rectangular cross-sectional modeling domain for the initial two-dimensional model of infiltration and tritium migration between the CAMBRIC ditch and monitoring well UE-5n. The well screen is 3 m long and is centered at a depth of 220 m, just below the water table. The curve indicates a hypothetical pathway for a parcel of tritium.

3.1 Model Conceptualization.

The rectangular cross-section shown in Figure 5 was chosen to develop a two-dimensional model of infiltration and tritium migration between the CAMBRIC ditch and monitoring well UE–5n. This model is used in conjunction with additional chemical isotope data to better analyze the history and nature of infiltration and radionuclide migration over the past 30 years. The model augments the previous effort of Ross and Wheatcraft (1994) in the sense that the tritium arrival data in UE-5n collected since 1994 can now be used for model calibration purposes, as well as to verify the utility of isotopic age dating techniques in saturated-unsaturated systems. In addition, several conceptual approximations required for numerical convergence in the previous model are no longer required. A calibrated model can then be used as a basis to estimate tritium flux across the water table, examine the longer term (> 30 y) fate of tritium and its daughter product (^3He) in the vadose zone, evaluate similar behavior for other isotopes observed in the ditch effluent, and plan future field measurements.

The location of the cross section is considered to be sufficiently far from the pumping well so that the assumption of two-dimensional behavior is adequate. Because the hydraulic gradients in the vicinity of CAMBRIC are close to 0.005 or smaller (Tompson, et al., 1999a), the water table has been assumed to be flat in this cross section, with a no-flow axis of symmetry underlying the center of the ditch. The water table is located at a depth of 214 m, consistent with observations in Well UE-5n (Ramspott and McArthur, 1977), and is over 80 m above the bottom of the domain, located at a depth of 300 m, that is also assumed to be a no flow boundary. The right-hand side of the domain was located some 400 m away from the ditch centerline in order that the water table elevation can be fixed there, at the same depth of 214 m, as a boundary condition. Water and tritium inflow to the system will be specified over a 1 m long half-width of the ditch, in the upper left-hand corner of the domain, and allowed to infiltrate towards the water table, accumulate as a mounded groundwater over the initial water table, and eventually exit the domain through the fixed head boundary on the right-hand side. It should also be mentioned that the hydraulic setting in the unsaturated media, above the water table on the right-hand side boundary, is also fixed or maintained in order to yield no-flow conditions. The USNT module of the NUFT computer code (Nitao, 1999) was used for the current water flow and tritium transport simulations.

Frenchman Flat is an intermountain basin formed by Tertiary-age faulting typical of the Basin and Range physiographic province. The model domain is located in Quaternary/Tertiary alluvium composed of interbedded silts, clays, sands and gravels derived largely from silicic volcanic rocks (tuff and rhyolitic lava). Reactive minerals such as clinoptilolite, calcite, smectite, illite, and iron oxide exist and will contribute some sorptive potential to the medium. For simplicity, the geologic system was treated as uniform in terms of its hydraulic properties even though it is clear that small-scale heterogeneity exists both within the modeling domain (Ramspott and McArthur, 1977; Tompson et al., 1999), and elsewhere in Frenchman Flat (REECO, 1994; Istok, et al., 1994; Blout et al., 1995).

3. 2 Transport and Constitutive Relationships.

Both liquid and gas phase flow (water and air) are considered in the NUFT model, along with appropriate interphase mass transfer processes. In general, the inclusion of a dynamic air phase and allowance for interphase mass exchange are pertinent issues when volatile species such as ^{85}Kr or ^3He are considered, but only of secondary importance with tritium. In this model, the flow of water is governed by mass and momentum balance equations (Nitao, 1999; Bodvarsson et al., 2000) given by

$$(1) \qquad \frac{\partial(\phi S_w \rho_w)}{\partial t} + \nabla \cdot \left(\phi S_w \rho_w \mathbf{v}_w\right) = e_{wa}$$

and a generalized form of Darcy's law,

$$(2) \qquad \phi S_w \mathbf{v}_w = -\mathbf{k} \cdot k_{rw}\left(\nabla p_w - \rho_w \mathbf{g}\right)$$

Here, $\mathbf{v}_w$ is the seepage velocity of water (L/T), S_w is the water phase saturation (dimensionless), and ϕ is the medium porosity (dimensionless). In (2), p_w is the water phase pressure (M/LT^2), ρ_w is the water density (M/L^3), $\mathbf{g}$ is the gravitational vector (L/T^2), and $\mathbf{k}$ and $k_{rw}(S_w)$ are the intrinsic permeability tensor and relative water phase permeability of the medium, respectively (L^2). For reference, the intrinsic permeability is related to the saturated hydraulic conductivity via $\mathbf{K}_s = \mathbf{k}\rho_w g / \mu_w$ (L/T) where μ_w is the water viscosity (M/LT) and $g = |\mathbf{g}|$. The quantity e_{wa} accounts for mass transfer between the air and water phases (M/L^{3}T), which will be negligible here.

The transport of tritium in water is governed most generally by

$$(3) \qquad \frac{\partial(\phi S_w c)}{\partial t} + \nabla \cdot \left(\phi S_w c \mathbf{v}_w\right) + \nabla \cdot \mathbf{J}_c = -r_{wa}$$

where c is the tritium abundance, $\mathbf{J}_c$ is the hydrodynamic dispersive flux, and r_{wa} is the loss rate of tritium from radioactive decay or interphase mass transfer. The tritium abundance may be expressed in any number of ways, such as an aqueous mole fraction (dimensionless), aqueous mass density (M/L^3), or radiochemical activity (Ci/L^3 or Bq/ L^3). Tritium exchange with the air phase is negligible, while radioactive decay is treated by correcting all tritium levels data back to their 1965 (zero time) values.

When the air phase (atmospheric) pressure (p_a) is assumed static, the water retention or capillary pressure constitutive function is represented as

$$(4) \qquad p_a - p_w = P_c = P_c(S_w)$$

where the Van Genuchten relationship (Nitao, 1999; Bodvarsson, et al., 2000) is used to parameterize this function, namely,

$$(5) \qquad P_c(S_w) = \frac{1}{\alpha^*} \cdot \left[S_w^{*\,-1/m} - 1\right]^{1-m}$$

Here, S_{wr} is the residual water saturation, $S_w^* = (S_w - S_{wr})/(1 - S_{wr})$, $\alpha^* = \alpha / \rho_w g$, and α and m are moisture retention parameters (L^{-1} and dimensionless, respectively). In (2), the relative permeability is modeled in terms of a complementary Van Genuchten relationship (Nitao, 1999; Bodvarsson, et al., 2000),

$$k_{rw}(S_w) = \left(S_w^*\right)^{1/2}\left[1 - \left[1 - \left(S_w^*\right)^{1/m}\right]^m\right]^2 \tag{6}$$

Thus, in addition to boundary and initial condition information, the parametric medium properties required for the flow model include the intrinsic permeability tensor, $\mathbf{k}$, the residual saturation, S_{wr}, and the moisture parameters α^* and m.

3.3 Model Application.

The model domain in Figure 5 was discretized into an orthogonal mesh of 81 by 136 grid blocks, with irregularly-sized cells ranging from 0.5 by 0.5 m underneath the ditch to 16 by 8 m near the bottom, right-hand side of the domain. The x-z coordinate system is anchored at the ditch centerline, with the *x*-axis increasing positively to the right in Figure 5, and the *z*-axis increasing positively down. The finest resolution of the grid is concentrated on the left-hand side of the domain, between the ditch centerline and monitoring well UE-5n. The upper row of blocks, with the exception of the left-most two, is treated as a dry ($S_w = 0$) atmospheric interface with a porosity of 0.99. Recharge from precipitation was not considered in this model. The left-most two blocks in the upper row, spanning a horizontal distance of 1 m (with an 0.5 m depth), represent one-half of the ditch width and were treated as ditch recharge cells.

Fixed water pressure conditions consistent with an approximate water depth of 2.5 cm (or about 1 inch) in the ditch were specified in the two ditch blocks as a means to force water to infiltrate into the system. Computed water fluxes across this interface were compared with the previously estimated influx rate, as a means to calibrate values assigned to the medium permeability. This approach did not fully represent the lower ditch flow rate in the first 700 days of pumping. Alternatively, the estimated influx rate could have been assigned directly as an inflow condition, although some adjustment would have to be made for the lower initial pumping rate, and the transient variation of the infiltration that would occur as the upper portions of the domain become increasingly saturated would not be reflected in this approach. Tritium migration was addressed by setting its activity in the ditch inflow to match the elution profile shown in Figure 4, but only though 1991, after which it was set to zero. Transpiration of tritium by phreatophyte vegetation surrounding the ditch, although suggested in the ditch flow observations and analyzed by Love et al. (2002), was not included in this model.

Because the geologic system is being treated as homogeneous, fixed parametric properties are specified at all locations in the domain, although the intrinsic permeability will be considered as an anisotropic quantity with principal axes aligned with the *x* and *z* model axes. This approach ignores small-scale infiltration and transport effects associated with fine-scale heterogeneity. It assumes that the traditional balance equations and constitutive relationships in (1-6) are valid at the larger field scale considered here (e.g., Mantoglou and Gelhar, 1987), despite the fact that most of the parametric measurements available in the literature are representative of medium behavior on relatively small scales (~0.1 to 1.0 m).

Table 1. Summary of hydraulic conductivity and soil retention data used in the representative flow simulations.

Run	Saturated vertical permeability, k_z (m^2)	Saturated horizontal permeability, k_x (m^2)	Anisotropy ratio, k_x / k_z	α^*(Pa^{-1})	m	Porosity, ϕ	Residual water content, S_{wr}
“ba”	6.10 e-13	4.88 e-12	8	1.02 e-3	0.45	0.33	0.20
“a”	5.50 e-13	4.95 e-12	9	1.02 e-3	0.52	0.33	0.20
“e”	4.44 e-13	4.00 e-12	9	1.02 e-3	0.60	0.33	0.20
“g”	4.00 e-13	3.60 e-12	9	1.02 e-3	0.60	0.33	0.20
“ga”	4.00 e-13	3.60 e-12	9	0.80 e-3	0.60	0.33	0.20
“h”	3.00 e-13	2.70 e-12	9	1.02 e-3	0.70	0.33	0.20
“j”	2.00 e-13	1.80 e-12	9	1.02 e-3	0.80	0.33	0.20

Hydraulic data for Frenchman Flat alluvium available in the literature were used to guide the development and approximate calibration of the model (Hoffman et al., 1977; Burbey and Wheatcraft, 1986; Istok et al., 1994; REECO, 1994; Ross and Wheatcraft, 1994). Parameters used in several representative simulations are shown in Table 1. Factors considered in this process involved:

- Setting permeability values within the reported ranges in the literature while using realistic anisotropy factors within the 2 to 10 range, so that the estimated infiltration rates underneath the ditch could be approximately matched while avoiding the creation of any significant mounding effect at the water table;
- Setting realistic values of α^* and m within the observed ranges;
- Fixing the porosity and background residual water saturation to uniform values of 0.33 and 0.20, respectively;
- Fixing the effective diffusivity of tritium at 10^{-9} m^2/s, while neglecting dispersion fluxes; and

Recognition that a completely homogeneous model, and one in which precipitation recharge and evapotranspiration are excluded, may not be wholly sufficient to match every observation at the site.

3.5 Model Results.

Figure 6 shows water saturation profiles at four different times since the initiation of pumping for simulation “g” (Table 1). Here we see a moisture plume sinking from the ditch and reaching the water table between 3 and 5 years. At $t = 25$ years, corresponding to the year 2000 – 9 years after the cessation of pumping – we still see a significant moisture plume in the system. Complete drainage back to ambient (residual) conditions will take a long time, and, in the model, is ultimately controlled by the permeability and soil moisture retention parameters. In Figure 7, the computed ditch inflow rate for this problem, corrected for the entire ditch width, is shown to vary between the estimates (0.01 to 0.02 kg-water/s per meter length of ditch) inferred from the flume-based flow measurements. The decrease in inflow rate over time is due to the increasing saturation conditions beneath the ditch, which will tend to constrain inflows as the moisture plume expands.

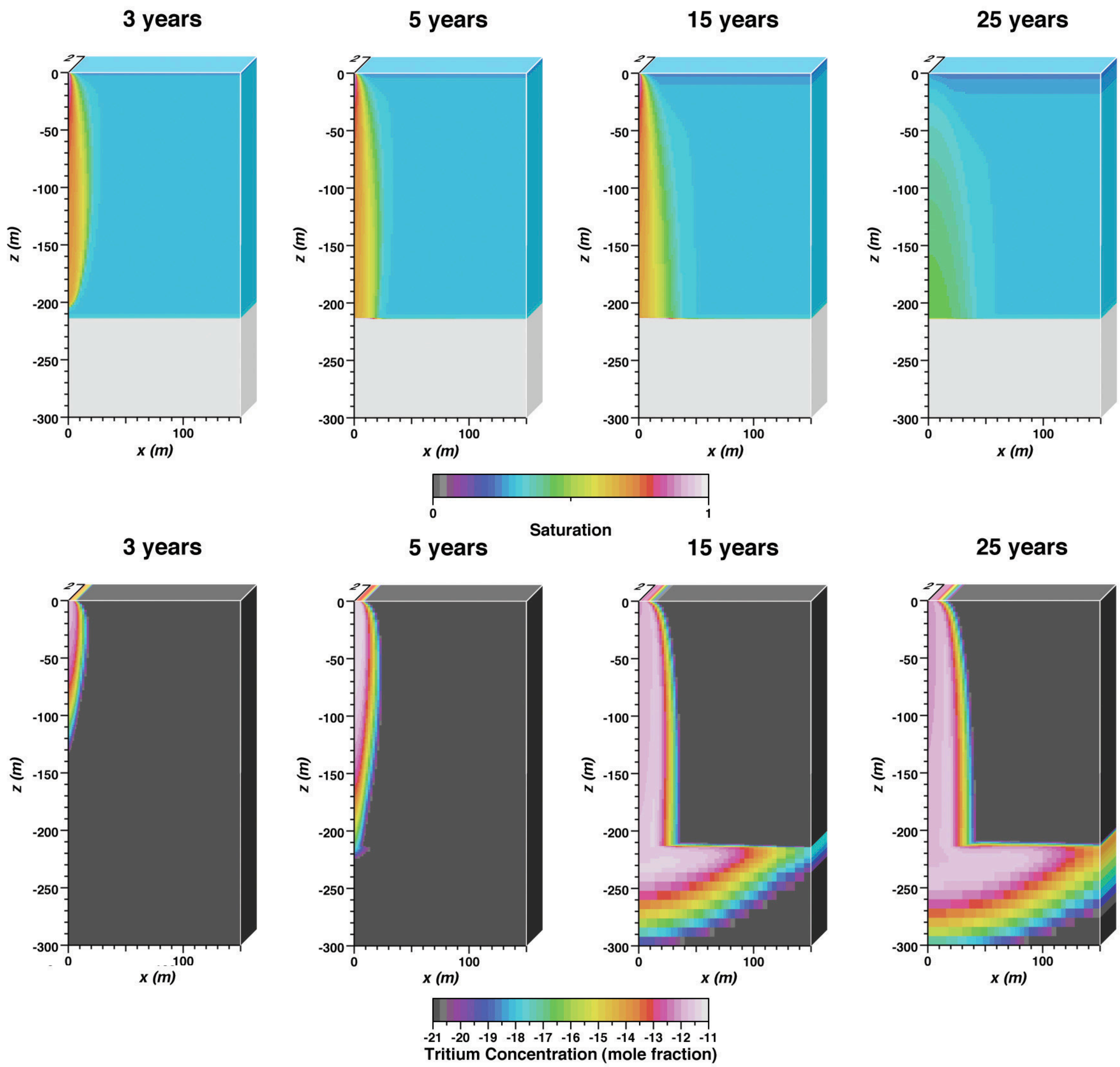

Figure 6. Liquid phase saturation (above) and tritium concentration (below) profiles predicted from simulation "g" (Table 1). Dates refer, approximately, to time after initiation of pumping in October 1975. Initial moisture arrival at water table is predicted between 3 and 5 years; initial tritium arrival is just about 5 years. Monitoring well UE-5n is located at x = 106 m. Tritium concentrations are decay corrected to May, 1965.

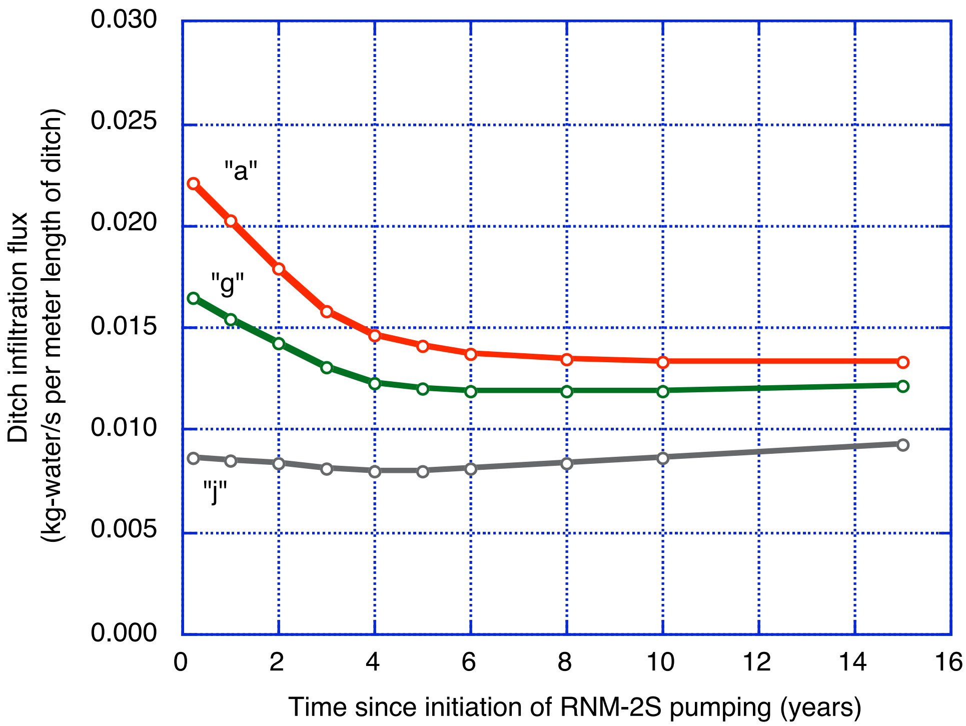

Figure 7. Computed ditch inflow rate, corrected for the entire ditch width, as a function of time for representative simulations "a", "g" and "j" of Table 1. The rates compare favorably with estimates (between 0.01 and 0.02 kg-water/s per meter length of ditch) inferred from the flume-based flow measurements.

The spatial distribution of tritium as a function of time determined in the "g" simulation is also shown in Figure 6, as expressed in terms of its mole fraction in the liquid water phase. The first arrival of tritium at the ditch occurs at about $t = 5$ years, which, when the approximately 2-year (900 day) delay in its release into the ditch is taken into account, suggests a slightly shorter travel time of about 2.5 to 3 years to the groundwater in the "established" moisture plume. After reaching the water table, the tritium is shown to migrate toward the UE-5n monitoring well (at x = 106 m), where the first arrival occurs just prior to the 15-year point.

The average activity in the well screen (near the water table), as computed by the model in simulation "g", is shown in Figure 8 and compares reasonably well with the historically measured data. Several other arrival profiles obtained from the additional simulations listed in Table 1 are also shown in Figure 8. These results correspond to slight variations in the values of permeability, m, and□*, and are quite indicative of the sensitivity of the model (and system conceptualization) to these parameters. Of particular interest is the sensitivity of the peak concentration and its timing to these parameters.

The results in Figures 6 and 8 clearly portray the longevity and widespread distribution of residual tritium in the unsaturated zone well after ditch flows have ceased. Figure 9 shows the computed tritium flux between the vadose zone and the water table, per unit length of the ditch, and as corrected to account for inputs over the entire ditch width, for simulation "g". This result is also indicative of the length of time tritium will remain in the vadose zone.

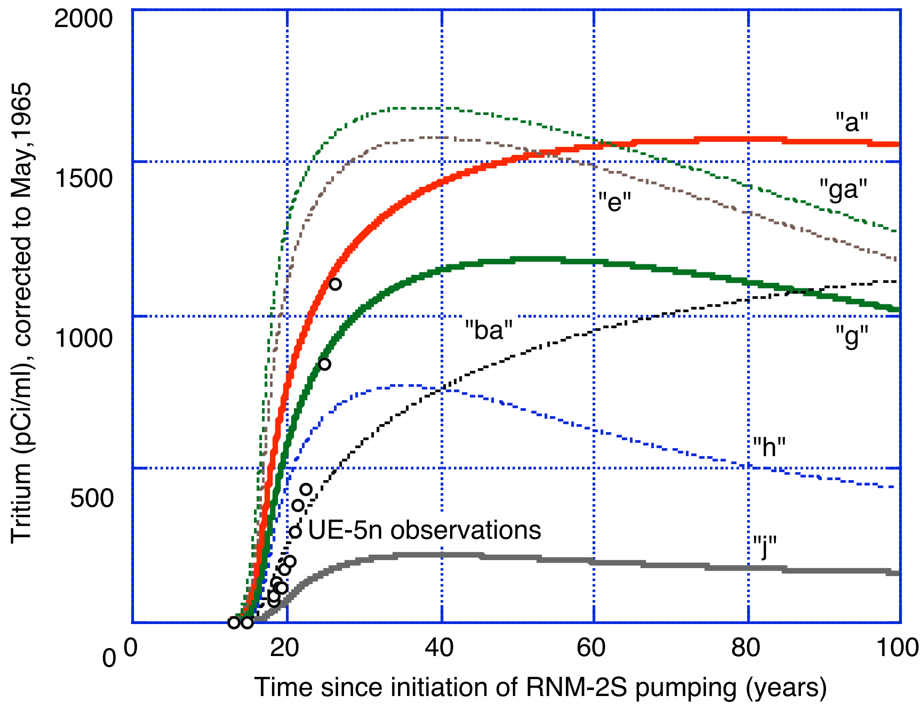

Figure 8. Predicted tritium activities in well UE-5n from the various simulations in Table 1, decay corrected to May 14, 1965, as compared with observations. Time scale refers to years since initiation of pumping in 1975.

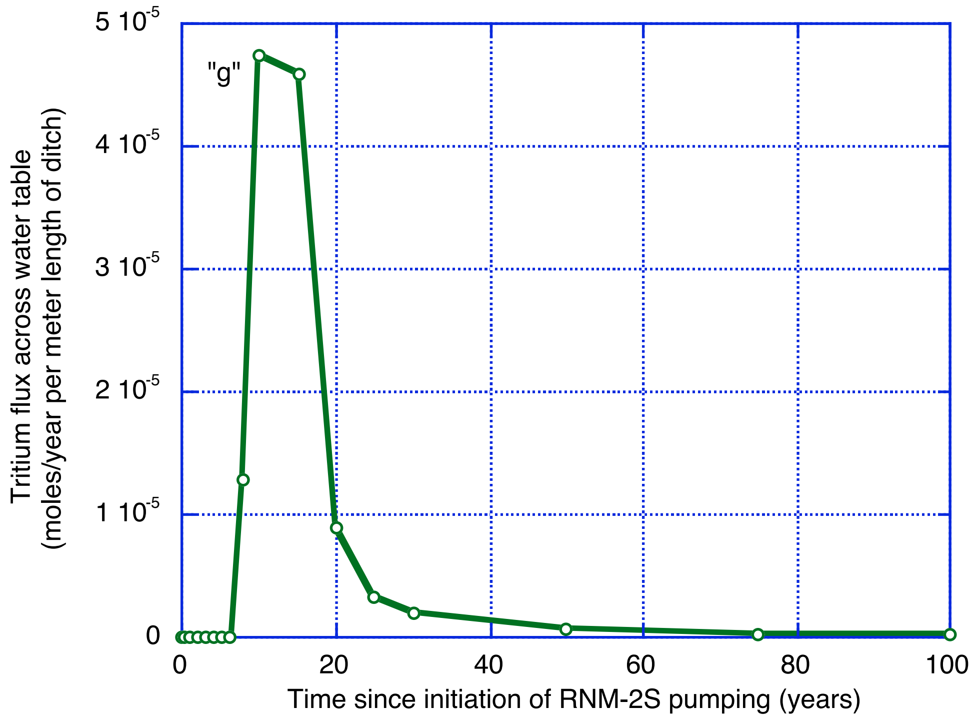

Figure 9. Predicted tritium flux across the water table as a function of time, per unit length of the ditch for simulation "g", and as corrected for inputs over the entire ditch width. Tritium fluxes are decay corrected to May, 1965.

4. New Chemical Measurements in the CAMBRIC System

Between 2000 and 2002, additional groundwater samples were collected from the pumping well (RNM-2S), the monitoring well (UE-5n), as well as the CAMBRIC cavity drillback hole (RNM-1, Figure 2). The samples were analyzed by mass spectrometry for tritium, $^{3}He/^{4}He$, ^{14}C, $^{36}Cl/Cl$, ^{85}Kr (by counting), ^{99}Tc, and ^{129}I, as well as for stable isotopes of D/H and $^{18}O/^{16}O$, but not for ^{106}Ru (Rose et al., 2002; Finnegan and Thompson, 2001).

Many of these isotopes are soluble and serve as excellent tracers of groundwater flow. From the new data, ^{14}C was identified with tritium in the pumping well, suggesting their co-migration from the test cavity during the pumping experiment and their joint introduction into the ditch. New data from the monitoring well indicated the presence of tritium, ^{36}Cl, ^{129}I,and ^{99}Tc, suggesting their co-migration during ditch infiltration, but no ^{14}C or ^{85}Kr, both of which were present in ditch water. This suggests, most likely, that ^{14}C interacted chemically with calcite in the alluvium, while ^{85}Kr partitioned into the atmosphere during flow in the ditch and soil gases during infiltration.

The tritium/^{3}He ratio can be used as a barometer of groundwater age, or travel time under saturated (below water table) conditions where the ^{3}He that accumulates from tritium decay is constrained to remain in solution (Tompson et al., 1999b). Measurements at the monitoring well UE-5n suggest a tritium age of 9.5 years. Provided that it took approximately 13 to 15 years for the initial tritium discharged into the ditch to reach UE-5n (see above), this suggests, in turn, a transit time through unsaturated conditions (i.e., the vadose zone) of 3.5 to 5.5 years. This result is in reasonable agreement with the transit time predicted by the simulations, indicated in Figure 6 above, and illustrates how age dating can be used as a tool to provide finer calibration of a saturated/unsaturated transport model.

The tritium age dating method was also applied to a new groundwater sample extracted from the original pumping well RNM-2S in 2000. In principal, this water should closely represent the last groundwater sample obtained from the well in 1991, at the end of the 16-year pumping period, largely because hydraulic gradients in this area are so flat that groundwater would not be expected to move very far under unstressed conditions. As this water has been wholly confined to saturated conditions, its age should reflect the time since the test – approximately 35 years. However, although the new tritium activity was similar to the most recent value obtained in 1991 (~ 1300 pCi/ml, decay corrected, Figure 4), the calculated age turned out to be only 20 years.

The simulations suggested a possible explanation for this discrepancy, namely, that tritium from the ditch, especially in its more upstream locations closest to the original pumping well, was recycled back to groundwater and recaptured by the pumping well. This concept was originally postulated by Ross and Wheatcraft (1994). When a recycled parcel of tritium returns to the water table, its apparent "age" will reset to zero because accumulated ^{3}He will partition preferentially into the atmosphere or soil gas during its transition into the ditch or through the vadose zone. Its mixing with "older", non-recycled tritium moving across the well screen will reduce the "apparent age" of the groundwater subsequently sampled from the well. This gas loss mechanism is also corroborated by the apparent loss of ^{85}Kr along flow pathways connecting the ditch to the monitoring well. The apparent dilution of age in the pumping well suggests that close to 60% of the tritium (and, by deduction, other mobile radionuclides) extracted from the well in 2000 was derived from recycled water. By extension, it would seem that a significant fraction of the extracted tritium indicated in Figure 4 has been recycled, especially in the latter years when the recirculation flows have probably become steady. Thus, it would appear inappropriate to wholly compare the integral of the recovery curve to some fraction of estimated CAMBRIC tritium inventory without considering recirculation effects (Hoffman et al., 1977; Guell and Hunt, 2003). Nor would it be correct to directly use the curve as an isolated calibration target for saturated zone models of tritium migration between the test cavity and pumping well (Burbey and Wheatcraft, 1996; Tompson et al., 1999a). The existence of recirculation is further supported by the data from the stable isotopes of $^{18}O/^{16}O$ and D/H that clearly show an enrichment in the heavier isotope that accompanies evaporation during water recycling (Figure 10).

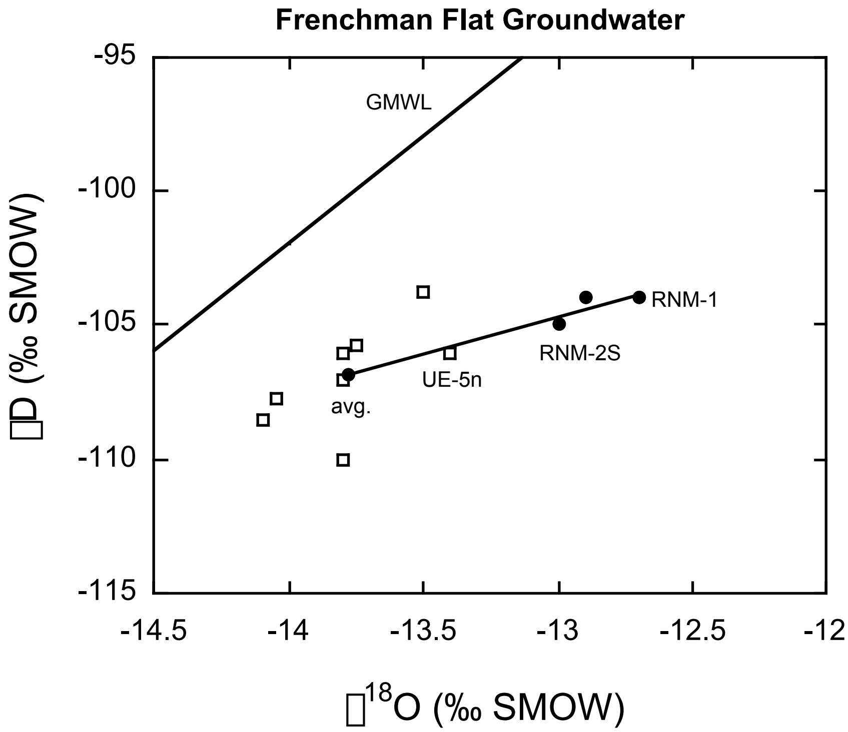

Figure 10. $^{18}O/^{16}O$ and D/H data from recent measurements in RNM-2S and RNM-1 (closed circle on right) that show an increased degree of evaporation relative to other measurements in nearby Frenchman flat groundwater wells (open squares). The global meteoric water line (GWML) is shown for comparison.

5. Conclusions and Recommendations

Altogether, the current simulations and chemical observations suggest that the transport of tritium, ^{36}Cl, ^{99}Tc, and ^{129}I through the 220 m vadose zone under the ditch discharge conditions is relatively fast (3 to 5 years). However, model simulations suggest their residence-time in the draining system following the shutoff of the ditch will be much longer in comparison.

Contrary to original perceptions, the role of the vadose zone on radionuclide transport under the flowing conditions of the ditch was non-trivial as evidenced by a significant return of mobile radionuclides to groundwater. The model can now be used to quantify the flux of radionuclides reaching the water table underneath the ditch, as in Figure 9, which is relevant to the protection of groundwater resources at NTS. We also conclude that the presence of radionuclides in UE-5n is entirely due to discharges from ditch proper, as opposed to transport in groundwater under ambient conditions from the nearby DILUTED WATERS or WISHBONE underground nuclear tests (Figure 3), both of which were detonated near the water table (USDOE, 2000).

The long-term nature of the CAMBRIC pumping and infiltration experiments has generated a unique opportunity to combine a variety of chemical and isotopic measurements made over the past 25 years into a more unified modeling analysis. In particular, the model simulations and chemical age dating results determined in this study here are in approximate agreement with respect to travel times between the ditch and monitoring well UE-5n. In addition, the existence of recirculation between the ditch and the pumping well will require a reevaluation of the original pump test data, as well as the myriad of models that have been applied to analyze radionuclide migration in the saturated zone between the CAMBRIC cavity and well RNM-2S. These have all attributed the entirety of the extracted radionuclides as coming directly from the test cavity without the influence of recirculation.

The vadose zone model results, although generally reasonable in many respects, were shown to be quite sensitive to the character and nature of the soil moisture and relative permeability relationships. This is, of course, typical of unsaturated flow models. The ability to match rising tritium levels in well UE-5n with the model was also, most likely, complicated by the presence and impacts of geologic heterogeneity.

All of the radionuclides detected in pumping well RNM-2S (tritium, ^{3}He, ^{14}C, ^{36}Cl, ^{85}Kr, ^{129}I, and ^{106}Ru) were able to move there from the CAMBRIC cavity during the pumping experiment and were, thus, relatively mobile under saturated conditions. However, in the ditch and the unsaturated zone beneath it, ^{3}He and ^{85}Kr preferentially partitioned into the gas phase, lowering their mobilities, while ^{14}C appears to have been retarded or retained from interactions with calcite minerals.

Although not directly incorporated in the model, the ditch infiltration rates were moderated by the transpiration processes of rooted vegetation, as evidenced by diurnal fluctuations in ditch flow rates and accumulations of tritium identified in the vegetation itself (Love et al., 2002). Preferential uptake of different radionuclides in the vegetation may also affect their apparent mobilities in the unsaturated zone.

5.1 Future Opportunities

Because of their 25-year long record of data, analysis, and observations, the CAMBRIC experiments still offer numerous opportunities for expanded modeling activities, additional data collection, and broader interdisciplinary syntheses to improve our understanding of deep vadose zone hydrology. Such opportunities include:

- Additional three-dimensional modeling studies to better (i) characterize the nature of recirculation between the upper extent of the ditch and pumping well RNM-2S, (ii) understand the influence of heterogeneity on moisture infiltration and chemical recirculation processes, and (iii) quantify the fate of all radionuclides introduced in the ditch, including their flux across the water table, transfer into the gas phase, or reaction with solid phase minerals.
- Lagrangian (or particle-based) simulations of tritium infiltration, ^{3}He generation, and ^{3}He fate as a means to examine additional details of the tritium age-dating technique in saturated/unsaturated systems and assess the utility of measuring of ^{3}He in soil gases as an additional way to characterize tritium migration;
- Continued collection and synthesis of radionuclide concentration and related groundwater quality data in Well UE-5n; and
- Development of new measurement and sampling opportunities over the full depth of the vadose zone, preferably in one or more new boreholes positioned close to the ditch along a perpendicular line connecting well UE-5n and the ditch. Such observation

locations could be used to collect alluvium samples for analysis of residual moisture, tritium, and other residual radionuclide content, such as ^{14}C, soil gas samples for analyses of ^{3}He generation and migration, or other biogeochemical samples that may reflect on the indigenous and altered ecological behavior in the subsurface resulting from the 16 years of infiltration.

- In addition, the ability of saltcedar and cattail shrubs surrounding the CAMBRIC ditch to bind tritium in their cellulose has recently been demonstrated by Love et al. (2002). The concentration of tritium in successive tree rings tracks the elution profile of tritium into the ditch from RNM-2S and may be exploited to more carefully quantify the transfer of infiltrated ditch water to the nearby vegetation (and its loss from the vadose zone).

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